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A theoretical study of the electronic structure of selected transition metal impurities in silicon carbide is carried out using a first-principles approach. Polytype dependence, site preference and charge and spin states are investigated.

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Research Progress Report

First-Principles Theory of Transition Metal Impurities in Silicon Carbide

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1. Personnel

The first task accomplished was to hire personnel for the project. A graduate student, Margarita Prikhodko started working on the project as of May 2000. She has finished her first year of graduate studies and passed the qualifying exam in August 2000. During the spring, as of January 2000, she was supported on a previous grant of ONR, and starting receiving training in the computational methods to be used for this project. During the summer, and fall, she worked on a first subproject related to the present grant.

A postdoctoral research associate, Dr. Maosheng Miao was hired as of October 1, 2000. After conducting a search for suitable candidates, an offer was made to him in April. However, his arrival was delayed by the time required to obtain an H1B visa. Dr. Maosheng has already completed several years of postdoctoral research in Belgium and came with excellent recommendations.

The delay in finding personnel is the main reason why negligible funds were spent during the first months of the project. We plan to make up for this delay by requesting a no-cost extension at the end of the project.

2. Scientific Research Overview

While the project got a somewhat delayed start due to lack of personnel, some preparatory work was accomplished in the spring. A new full-potential linear muffin-tin orbital method was tested for SiC. This new method, developed by Michael Methfessel and Mark van Schilfgaarde uses smoothed Hankel functions as envelope functions of the basis set, which reduces the size of the basis set. It also includes the capability of calculating the forces analytically and an automatic procedure for relaxing the atomic positions is implemented in the method using a conjugate gradient approach. This feature is important to relax the structure around defects in solids. Before applying the method to defects in SiC, we decided to test its accuracy for some bulk properties of SiC. In particular, we tested how it performs on elastic constants and phonon frequencies. An optimized basis set and related convergence parameters (mesh size, angular momentum cut-offs, k-point set) providing accurate results for these properties was determined. A table of some of these results comparing to previous results and experiment is provided below.

Table I: Calculated properties of 3C-SiC

Property	*Present*	*Other method**	*Experiment*
Lattice constant a (Å)	4.33	4.315	4.36
Bulk Modulus B (GPa)	231	223	225
C_{11} (GPa)	403	420	390
C_{12} (GPa)	145	126	142
C_{14} (GPa)	253	287	256
Kleinman ζ	0.41	0.49	
ω_{TO} (cm^{-1})	798	796	796

* Other method refers to older FP-LMTO approach W. R. L. Lambrecht, B. Segall, M. Methfessel, M. van Schilfgaarde, Phys. Rev. B **44**, 3685-3694 (1991) in which also the experimental values are discussed and their original references provided.

By participating in these calculations, the graduate student was instructed on the basics of the density functional method and linear muffin-tin orbital methods, including the new full-potential version and learned how to use the related computer programs. During the summer she conducted the first tests on defect calculations. The C vacancy in cubic 3C-SiC was used as a test case because of the availability of prior results in the literature.[Zywietz] This paper provides detailed results on the relaxations as well as energy levels which we found to agree well with our preliminary calculations.

As a further exercise for the student in the use of the new method, and to gain more experience with relaxation calculations before we start on the large unit cell defect calculations, we then embarked on a study of the high-pressure phase transition of SiC from the zincblende to the rocksalt structure. This work was inspired by results obtained on the GaN wurtzite to rocksalt transition, accomplished under a prior ONR grant (No. N00014-98-1-0160) and which has led to a paper recently accepted for publication in Physical Review Letters.[Limpijumnong] This addresses an old problem in solid state physics: namely the preference for tetrahedral versus octahedral bonding. The new insights we obtained pertain primarily to the mechanism underlying this phase transition. Whereas previously it was thought that softening of acoustic phonons plays the primary role, we realized that the acoustic phonon softening indirectly leads to an instability in the optical phonon mode via the internal strain coupling. In other words, the interaction between strain and the relative position of the two sublattices leads to the phase transition instability. The zincblende structure can be viewed as two interpenetrating fcc lattices displaced by **u**=(1/4,1/4,1/4) whereas the rocksalt structure has a relative displacement of **u**=(1/2,1/2,1/2). Displacements of the atoms along the [111] direction correspond to the optical phonon vibrational mode. We calculated the energy as function of **u** between these two limiting values for various values of the lattice constant and additional uniaxial traceless compressive strains along the [111] direction. The preliminary results are that whereas for the equilibrium lattice constant, there is only a single minimum as functon of **u** at the zinblende value, a second minimum develops near the rocksalt value of u=1/2, and eventually obtains a lower energy than the zinblende one. The barrier between the two minima is significant and in fact increases with decreasing lattice constant. This barrier, however, appears to be decreasing under additional [111] strain. This indicates that strain in the [111] direction, which can be viewed as the long-wavelength limit of acoustic phonons in the [111] direction facilitates the transition from zincblende to rocksalt. This work will be presented at the upcoming APS March Meeting in Seattle. We also found that the equilibrium phase transition pressure is around 56 GPa in good agreement with a previous calculation reporting 60 GPa [Christensen] but much lower than the experimentally reported value of about 100 GPa [Yoshida]. This is indicative of slow kinetics in the experiments in agreement with our finding of a significant barrier.

Meanwhile, further work is in progress on the point defects. Further calculations on the vacancy, including charged defect states, and initial calculations on vanadium in 3C-SiC were started. Prior to this, the basis set for vanadium was optimized by performing calculations for bulk vanadium metal. Initial indications are that the relaxation around a substitutional V atom is significant.

An important problem for dealing with semiconductor defect levels is the well-known underestimate of the band gap by the local density approximation. This makes it difficult to ascertain the precise positions of defect energy levels versus the band edges. A promising new method to overcome this problem is the so-called Screened Exchange method. During the spring, some progress was made towards implementing this method. In particular, some deficiencies of a prior treatment in the literature were identified. These pertain to the treatment of the intra-atomic parts of the screened exchange. Work is in progress to implement this method in the context of the linear muffin-tin orbital approach. We anticipate that it will lead to more accurate defect calculations in the future.

3. Publications and Presentations

Computational study of the zincblende to rocksalt transformation in silicon carbide, Margarita Prikhodko and Walter R. L. Lambrecht, abstract submitted to the APS March Meeting, Seattle 2000.

References:

[Zywietz] A. Zywietz, J. Furthmüller, and F. Bechstedt, Phys. Rev. B **59**, 15166-15180 (1999), Vacancies in SiC: Influence of Jahn-Teller distortions, spin effects, and crystal structure

[Limpijumnong] S. Limpijumnong and W. R. L. Lambrecht, Phys. Rev. Letters (2000), accepted, Homogeneous strain deformation path for the wurtzite to rocksalt high-pressure phase transition in GaN

[Christensen] N. E. Christensen, S. Satpathy and Z. Pawlowska, Phys. Rev. B **36**, 1032-1050 (1987) Bonding and ionicity in semiconductors

[Yoshida] M. Yoshida, A. Onodera, M. Ueno, Phys. Rev. B **48**, 10587-10590 (1993), Pressure induced phase transition in SiC